Revised Draft

EVALUATION OF RADON LEVELS IN BUILDING 40 AT BLOCKSON CHEMICAL

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SUMMARY

At the request of the Advisory Board, SC&A presents this report to assist the Board in its deliberations on the SEC petition for Blockson Chemical Company. Described are multiple approaches for defining the bounding average annual radon concentration for Building 40 at Blockson during the qualification period.

SC&A first evaluated the application of the use of Florida phosphate industry data, as adapted from ORAUT-OTIB-0043, as a surragate for Blockson radon exposures. In this evalaution, SC&A found these data to be applicable in certain aspects and deficient in others. One deficiency that we identified in our earlier investigations had to do with the statistical analysis of surrogate data. SC&A re-analyzed the data used in OTIB-0043, extracting the individual analyses, where possible, from datasets used for the OTIB-0043 analysis. The bounding (95^{th} percentile) value assigned from OTIB-0043 was 2.33 pCi/L (0.112 WLM/y). After expanding the data, SC&A derived a 95^{th} percentile (bounding) radon concentration of 7.78 pCi/L. However, subsequent discussion of this matter with NIOSH revealed that NIOSH deliberately used the 95^{th} percentile value of the means of the datasets, as opposed to the 95^{th} percentile of the individual measurements, because the latter approach would be unrealistically conservative. SC&A agreed with this rationale, but suggested that this rationale be provided in OTIB-0043.

SC&A found on the "O" drive the results of a 1983 industrial hygiene survey performed at the former Blockson Chemical site by the (then) current owner, Olin Corporation (Marseglia 1983). At the time of the survey, the plant was continuing to operate as a phosphate products producer. The products made by Olin were fertilizers, rather than monosodium phosphate that was produced by Blockson Chemical. However, the phosphoric acid production, an intermediate step, was most likely not affected by this transition to different end products. In the 1983 survey, radon daughter measurements were made in three locations, which appear by their description to be in Building 40. For two of the locations, the radon results were nil; but at the third location, "40 filtration," by applying information from the report and other sources, SC&A found the WL and WLM from this survey to be 0.002WL and 0.0235 WLM/y, respectively, or less than 1 pCi/L.

SC&A went to great lengths, including additional worker interviews, with the participation of NIOSH and the chair of the work group, to evalaute the degree to which the measurements made in 1983 are indicative of those radon levels that might have been present during the qualification period. Our investigations revealed that there were most likely significant modifications made to the building ventilation systems between the qualification period and 1983. There is also evidence from worker interviews that the phosphoric acid production increased during this time frame, creating the potential for even greater radon concentrations in Building 40 in 1983, as compared to the qualification period. Nevertheless, we found it difficult to conclude that the radon measurements made in 1983 can be considered representative or bounding of the radon concentrations present during the qualification period.

Lastly, as a means to independently assess the reasonableness of NIOSH's default radon concentration of 2.33 pCi/L, SC&A performed a series of deterministic and probabilistic analyses to evaluate the possible radon concentrations that might have been present in Building

40 during the qualification period. In order to scope the range of potential radon concentrations, SC&A varied the radon release fraction from the ore dissolved in the acid tanks and the building ventilation rate to evaluate the possible range of concentrations. The following table summarizes the results of these deterministic calculations.

Table S-1. Radon Concentrations (pCi/L) based on Deterministic Analysis

ACH[a]	Release fraction from acid (f)					
	0.0	**0.2**	**0.4**	**0.6**	**0.8**	**1.0**
0.5	0.44	18.52	36.61	54.70	72.79	90.88
1.5	0.15	6.24	12.33	18.42	24.51	30.60
2.5	0.09	3.75	7.41	11.07	14.73	18.39
3.5	0.06	2.68	5.30	7.92	10.53	13.15
4.5	0.05	2.09	4.12	6.16	8.20	10.23
5.5	0.04	1.71	3.37	5.04	6.71	8.37

Note: A description of the models used to derive these values is provided in Appendix B.

[a] Air changes per hour

The term "deterministic" is used here, because the calculations are performed using a set of fixed or deterministic values for the key calculational parameters. As may be noted, depending on the values selected for the radon release fraction and air exchange rate, radon concentrations from below 1 pCi/L to as high as 90 pCi/L are derived.

In light of these uncertainties, SC&A performed a Monte Carlo simulation of the model, which we believe can help the Board in judging which of these values best represents the radon concentration in Building 40 during the qualification period. The results of the Monte Carlo simulation are as follows.

Table S-2. Percentile Values of ^{222}Rn Concentrations (pCi/L)

Percentile	Value
0%	0.02
5%	0.78
10%	1.50
15%	2.24
20%	2.97
25%	3.70
30%	4.44
35%	5.20
40%	5.99
45%	6.84
50%	7.72
55%	8.73
60%	9.87
65%	11.28
70%	13.06
75%	15.54
80%	19.08
85%	24.77
90%	35.37
95%	61.95
100%	651.00

The results indicate that the default value of 2.33 pCi/L selected by NIOSH in OTIB-0043 falls within the range of values and may, in fact, be an appropriate value, especially if only a small fraction of the radon in the ore entering Building 40 escapes from the ore during the grinding and digestion process and enters the Building 40 atmosphere. However, given the large uncertainties in radon release fractions from the ore during crushing and digestion and the uncertainty in the air exchange rate for Building 40, a higher default value may be needed. For example, the results of this analysis indicate that one can be 95% confident that the average airborne radon concentration in Building 40 during the qualification period was less than about 62 pCi/L.

1.0 BACKGROUND

On January 10, 2007, SC&A issued its review of the Blockson Chemical Company Special Exposure Cohort Petition (SEC), the Technical Basis Document (TBD) (Tomes and Glover 2007), the Evaluation Report, and selected supporting documentation. One of the findings (Finding No. 5) in SC&A's draft report dealt with the data, models, and assumptions used by NIOSH for reconstructing radon exposures to workers at Blockson. Subsequent to the issuance of that report, there were numerous work group meetings and an exchange of white papers related to this matter, which were effective in addressing many aspects of this issue. This matter was further discussed at the Advisory Board meeting held in St. Louis on June 24–26, 2008. During that meeting, the Board requested that SC&A specifically address the results of a series of worker interviews and calculations that were performed by SC&A and NIOSH in the 2-week period prior to the meeting to gain further insight into the radon issue. This report has been prepared in response to that request, and to capture in one place the work group discussions that led up to the matters discussed at the St. Louis meeting.

The root cause of the radon issue at Blockson is that there are no identified measurements of radon at Blockson Chemical during the SEC qualification period of January 1, 1951, to December 31, 1962. The only available measurements were made during an industrial hygiene survey conducted in 1983 by the Olin Corporation, the then owner of the Blockson site (Marseglia 1983). These measurements were made well after the cessation of uranium extraction activities conducted in Building 55 and consisted of 10 radon working level (WL) measurements in various indoor and outdoor locations at the site. Although these data are of interest, there is some question as to whether they can be used to reconstruct radon exposures of workers decades earlier. To remedy this limitation, Tomes and Glover (2007) used radon measurements collected at other phosphate facilities (i.e., surrogate data) as the basis for reconstructing radon exposures of Blockson workers during the qualification period. Hence, there is some question whether the surrogate data can reasonably be used to bound the exposures at Blockson during this period. The following sections explore this issue.

2.0 APPLICABILITY OF ORAUT-OTIB-0043

Tomes and Glover (2007) and the SEC-00058 evaluation used the radon analysis from ORAUT 2006 to assign a bounding radon exposure for workers at Blockson. The surrogate data in ORAUT 2006 upon which the Blockson bounding radon exposure is based are from a report by Birky et al. (1998) and consist of measurements made in a number of phosphate plants in Florida. The plants employ the wet production process for phosphoric acid, similar to the

process at Blockson, and the radon measurements presented were made at various locations throughout the plant. Those measurements made in locations obviously not representative of phosphoric acid production, e.g., mining rock tunnels, wet rock loading, and mining operations, were appropriately excluded from the dataset.

The radon data listed by ORAUT 2006 were obtained using a number of measurement methods, including e-perm, track-etch, and continuous working level monitors. While all are recognized and trusted methods, each method has its unique quality factors, and no quality control data are presented upon which to determine the overall quality of the dataset. However, all these devices were included in the device performance testing portion of the former EPA Radon Proficiency Program. The suppliers of these devices were required to maintain quality assurance programs and meet rigid quality performance requirements as part of the testing program. As a result, SC&A concluded that the measurements met the data quality requirements required by the EPA.

In the analysis of the Birky data, ORAUT states that 130 data values are contained in the dataset; however, Attachment B of ORAUT 2006 lists only 128 values, including 7 values that are below the limit of detection (LOD). These values represent individual measurements as well as mean values of measurements that span periods of up to 14 years. According to ORAUT, "The datasets that gave summary radon concentrations for a span of years were given equal weight with those that gave concentrations over shorter periods." The variance of a set of values that include the mean values of a large number of individual measurements would be smaller than the variance of the individual values. Consequently, as discussed further in Appendix A to the present report, the 95th percentile, which ORAUT derives from this set of mean values, would be more indicative of the 95th percentile of the mean values than of the 95th percentile of the individual measurements.

Table B-3 of ORAUT 2006 lists summary statistics on measured radon concentrations at eight locations in the area of a chemical plant over a period of 5 years, including the mean, the sample variance, and the total number of samples at each location. These are unlike the data on the other 120 measurements, which comprise only the mean concentrations, even for measurements that span periods of up to 14 years, as well as individual measurements. SC&A calculated a weighted mean that represents 713 values, combining the mean values at these eight locations, which comprise 593 individual measurements, with the 120 other measurements, which, for lack of better information, were each assigned a weight of 1. This weighted mean was 2.08 pCi/L, compared to an unweighted mean of 128 values of 1.01 pCi/L. We also calculated an expanded variance of the 713 values. Based on this variance, we derived a 95th percentile value of 7.78 pCi/L, which is considerably higher than the 95th percentile value of 2.33 pCi/L (0.112 WLM/y) cited by ORAUT and adopted by Tomes and Glover (2007) for use in the assessment of Blockson workers.

SC&A suggested that a default radon concentration of 7.78 pCi/L might be a more appropriate value for Blockson Chemical for all locations covered in the SEC, except in the rock crushing/calciner area. In addition, we suggested an indoor equilibrium factor of 0.4, as recommended by UNSCEAR (2000). For the rock crushing/calciner area, SC&A suggested an outdoor equilibrium factor of 0.6, as recommended by UNSCEAR. Thus, SC&A suggested a revised (bounding) value for the rock crushing/calciner area of 0.56 WLM/y, and for all other

covered areas, we suggested a value of 0.37 WLM/y. A detailed discussion of the SC&A assessment of the radon data from ORAUT (2006) is presented in Appendix A.

These suggestions were provided to the work group in an SC&A white paper and discussed during a work group meeting held on June 5, 2008. During that meeting, NIOSH acknowledged that the recommended default value of 2.33 pCi/L of radon is more representative of the 95th percentile of a set of means of individual measurements than the 95th percentile of a set of individual measurements. However, NIOSH provided convincing arguments that, since no one individual would be expected to be continually exposed to the upper 95th percentile value, it is more appropriate to use the 95th percentile of the means as opposed to the 95th percentile of the individual values. SC&A concurred with this philosophy, but stated that ORAUT 2006 would benefit from a thorough discussion of this matter.

In addition to issues related to how the Birky et al. data were used, SC&A expressed concern regarding how well the data taken at wet process phosphate plants represent conditions at Blockson during the time period of uranium extraction. Given that we have only general descriptions of the Blockson monosodium phosphate production plant that we can compare to the Florida phosphate plants, we can only speculate that they were not vastly different in terms of the radon exposures that plant personnel experienced. SC&A's investigations into this matter in support of earlier work group meetings revealed that measurements in the Birky et al. dataset appear to represent activities at Blockson through phosphoric acid production, except that no radon measurements were made at calciner locations. Since most of the radium was removed in the production of phosphoric acid, it is not likely that significantly elevated radon levels would result from production of monosodium phosphate or the extraction of uranium.

EPA (1978) reported measurements of radon in the calciner area at wet process phosphate plants. The EPA study was performed at an Idaho wet process plant, where the radon level in the calciner was reported as 0.18 pCi/L of radon. This is equivalent to 0.0011 WL and 0.013 WLM/y. SC&A concluded that these low levels will have no affect on the analysis presented in OTIB-0043. It is noteworthy that it has been established through worker interviews that the calciner at Blockson was located outdoors. While it is not known if the calciner at the Idaho plant was indoors or outdoors, in either case it would not underestimate the associated values at Blockson.

One measure of the appropriateness of using Florida wet process plant data as a surrogate for Blockson is to compare the amount of ore processed at each of the facilities. The following comparison, which is based on an earlier SC&A white paper, assumes that the amount of phosphoric acid produced per ton of ore is equivalent at the two facilities, and the concentrations of radioisotopes in the phosphate ore, particularly ^{226}Ra, are equivalent. Since Blockson processed phosphate ore from Florida and the wet process for phosphoric acid production is used in both cases, these assumptions are reasonable.

Guimond (1977) states, "Since approximately 4 metric tons of gypsum are produced per ton of phosphoric acid, a large phosphoric acid plant would produce about 2.5 million metric tons of gypsum per year." Guimond credits Slack 1968 for the numbers. From this we can calculate that 6.25×10^5 tons of phosphoric acid is produced per year in a large phosphoric acid plant:

$$6.25 \times 10^5 \text{ (tons/y of phosphoric acid)} = \frac{2.5 \times 10^6 \text{ (metric tons gypsum/y)}}{4 \text{ (metric tons gypsum/ton acid)}}$$

In an OCAS response to a review by SC&A (OCAS 2007), Tom Tomes indicated that 100 lbs of U_3O_8 were produced per day by Blockson, and that each liter of phosphoric acid contained 63 mg of U_3O_8. Using these values, 7.2×10^5 liters of phosphoric acid would have been produced each day at Blockson:

$$7.2 \times 10^5 \text{ (L/day)} = [100 \text{ (lb/day } U_3O_8)][4.536 \times 10^5 \text{ (mg/lb)}] \div [63(\text{mg } U_3O_8 \text{ per L of acid})]$$

This is equivalent to 1.33×10^3 tons of phosphoric acid per day or 4.65×10^5 tons of phosphoric acid per year produced at Blockson.

$$1.33 \times 10^5 \text{ (tons/day)} = [7.2 \times 10^5 \text{ (L/day)}][1675 \text{ (g/L)}] \div [9.072 \times 10^5 \text{ (gm/ton)}]$$
$$4.65 \times 10^5 \text{ (tons/y)} = [1.33 \times 10^5 \text{ (tons/day)}][350 \text{ (days/y)}]$$

Thus the amount of phosphoric acid produced at a large wet-process phosphoric acid plant is 1.34 times greater than that produced per year at Blockson.

$$1.34 = \frac{6.25 \times 10^5 \text{ tons/y of phosphoric acid at a large plant}}{4.65 \times 10^5 \text{ tons/y of phosphoric acid at Blockson}}$$

The phosphate production at Blockson was similar in most respects to the wet process plants that were the basis of the ORAUT (2006) radon data. Given that the amount of ore, and presumably the annual inventory of ^{226}Ra available for producing radon, was less than that at larger plants, we were inclined to believe that the bounding radon values (perhaps as revised to accommodate the 95th percentile issue discussed above) presented by Tomes and Glover (2007) are acceptably representative of Blockson.

Since there are no measurements available before the mid- to late-1970s from phosphate plants, it is not possible to compare radon levels from the two time periods. As reported in one of SC&A's earlier white papers, based on available information (Birky 2005), SC&A then believed that the plant processes did not change significantly over the time frames involved, and that there were few, if any, process modifications instituted in the plants to reduce radon levels. There were personnel protective measures (e.g., the wearing of dust masks) and personnel monitoring (e.g., TLD monitoring) instituted at some phosphate plants. SC&A concluded at that time that the use of the later data, as described in OTIB-0043, especially the 95th percentile data (perhaps as revised), represents the earlier exposure conditions at Blockson in a sufficiently claimant-favorable fashion. However, the work group requested that SC&A and NIOSH explore this matter further.

SC&A further examined the appropriateness of the use of surrogate data obtained from the Florida phosphate industry for the Blockson process, specifically to determine if the phosphoric acid production plants in Florida were better ventilated than Building 40 at Blockson. From a review of the available literature, it appears that in most of the phosphate plants in Florida, the size reduction (ball milling) and the reactor (attack) tanks, where the pulverized phosphate rock was reacted with sulfuric acid, were in essentially open buildings with no external walls.

SC&A interviewed, via telephone, J. Wesley Nall, a member of the Polk County (Florida) Public Health Unit, who was one of the authors of the Birky et al. (1998) study of radiation exposures in the phosphate industry. He was quite familiar with phosphoric acid production plants and confirmed that they were housed in multi-story, mostly open-sided buildings with enclosed, air-conditioned control rooms. The flooring was steel grating, and no forced ventilation was used.

SC&A also spoke with a retired EPA employee who was involved in the studies of the phosphate industry in the 1970s. His description of the phosphoric acid plants was virtually identical to that described in the preceding paragraph.

These statements are based on generalizations of the Florida phosphate plants and, because the plants whose data were included in the Birky et al. (1998) dataset are not specifically identified, it is not possible to determine the exact nature of the enclosures for those phosphoric acid operations. However, based on all the information available on Blockson, including recent worker interviews, Building 40 contained the milling equipment and reactor tanks in an enclosed building with an unknown (if any) amount of forced ventilation, and it cannot be ruled out that the Florida plants were in open, better ventilated structures. Hence, at this point in our investigations, we were concerned that the surrogate data might have substantial limitations as applied to Blockson. This concern became the focal point of subsequent investigations by both NIOSH and SC&A, which are described below.

3.0 RADON MEASUREMENTS IN BUILDING 40

In 1983, the Olin Corporation, then the owner of the Blockson site, performed an industrial hygiene survey (Marseglia 1983). At the time of the survey, the plant was continuing to produce phosphate products. The products produced by Olin were fertilizers, rather than monosodium phosphate that was produced by Blockson Chemical. However, the phosphoric acid production was most likely not affected by this transition to different end products. The production rate of phosphoric acid at the time this survey was performed, relative to that at Blockson during the uranium extraction period, is not known, except in a general sense. Recent interviews (see "O" drive) of Blockson workers indicate that a significant increase in acid production occurred after the qualification period and before 1983. These same workers also indicated that some building/process modifications were instituted to exhaust dust and acid fumes.

During the 1983 survey, radon daughter measurements were made in three locations, which appear by their description to be in Building 40, the phosphoric acid production building. These are identified as, "40 grinder platform," "40 screening platform," and "40 filtration" (Marseglia 1983). The information contained in the report indicates the radon daughter measurements were made using the Kusnetz method. This method employs a high-volume air sampler to collect radon daughters and, following a measured time period, determines the gross alpha count rate. The radon concentration is then determined by applying the following formula (Birky et al. 1998):

$$WL = \frac{C}{K\ V\ E}$$

where:

WL = radon concentration in units of working level (WL)
C = net alpha count rate (gross alpha cpm - bkgd cpm)
V = volume of air sampled in liters (avg cfm × time sampled [min] × 28.32 L/cf)
E = counting efficiency (cpm/dpm)
K = correction factor as supplied on the "Kusnetz Method Working Level Sampling" form

The Kusnetz correction factor (K) is a predetermined factor whose value depends on the time lapse from sample collection to counting.

The following table, taken from the Olin survey report (Marseglia 1983), gives the locations and raw data for the radon daughter measurements.

Table 1. Radon Daughter Sample Results

Sample ID	Location	Sample Volume, Liters	Elapsed time, min	Counts/ 5 min
RAD-4-28-83-1	hygrade area	16.4	51	0
" 2	#32 belt area	16.4	49	4
" 3	40 grinder platform	16.4	86	0
" 4	40 screening platform	16.4	84	0
" 5	Gyp pile	30.1	52	5
" 6	SF	30.1	40	1
" 7	STPP	30.1	44	18
" 8	40 filtration	27.4	50	7
" 9	SSF	24.7	40	2
" 10	TRP-2 Northeast corner (downwind)	27.4	48	0

As can be seen in the above table, two of the measurements in Building 40 resulted in zero counts and one, "40 filtration," 7 counts per 5 minutes. Since we do not know the counter efficiency, we cannot use the above formula to directly calculate the WL value associated with 7 counts per 5 minutes. However, since the report gives a WL of 0.0042 for the STPP location (18 c/5min), the WL at "40 filtration" can be determined by:

$$WL_{40} = \frac{C_{40}\, K_{STPP}\, V_{STPP}\, WL_{STPP}}{C_{STPP}\, K_{40}\, V_{40}}$$

where the subscript "40" refers to the "40 filtration" location, while "STPP" refers to the "STPP" location.

WL_{40} = 0.00196 WL
C_{40} = 7
K_{STPP} = 142 (corresponding to elapsed time of 44 min) (Birky et al. 1998, Table B-7)
V_{STPP} = 30.1 L
WL_{STPP} = 0.0042 WL
C_{STPP} = 18
K_{40} = 150 (corresponding to elapsed time of 50 min) (Birky et al. 1998, Table B-7)
V_{40} = 27.4 L

This WL value measured in Building 40 in 1983 is well below the bounding WL value(s) recommended for Blockson. As such, these data would seem to confirm that the use of the Florida data as a surrogate for Blockson appear to be claimant favorable. However, the use of these measurements to discern the possible radon concentrations in Building 40 during the qualification period of January 1, 1951, to December 31, 1962, obviously depends on the degree to which the operations and conditions affecting radon concentrations were similar for the two time periods (i.e., 1951–1962 versus 1983). Based on worker interviews, there were changes to Building 40 to improve the ventilation and/or exhaust systems. One of the workers recently interviewed believed these changes took place in 1979 or 1980. Another recently interviewed worker believed that the ventilation was upgraded around 1961. In the late 1960s or early 1970s, plastic cones were placed over the digester tanks to improve ventilation. Both workers reported the production was increased during the terms of their employment, which started in 1947 and 1951, respectively. Hence, at this point in our investigations, we had concerns regarding the usefulness of both the Florida data and the 1983 Blockson data as surrogates for evaluating worker exposures to radon and radon progeny at Blockson during the qualification period.

4.0 BUILDING 40 RADON MODELING

Given the uncertainties discussed above regarding the assignment of bounding radon concentrations during the qualification period for Blockson Building 40, SC&A, together with NIOSH, initiated a series of telephone interviews with workers to gain additional insight into the Blockson operations and building layout, including ventilation, both during the qualification period and in 1983, the time at which the radon progeny measurements were made. This was done to determine if substantial changes in the ventilation system and operations might have occurred between these two time periods, changes that could help to confirm or possibly invalidate the use of the 1983 data as a surrogate for the qualification time period. During these inquiries, both SC&A and NIOSH realized that we were compiling information that would allow us to model the radon concentrations that may have existed in Building 40 during the qualification period. Hence, SC&A initiated a modeling effort approximately 1 week before the St. Louis meeting. As will be discussed, the modeling effort proceeded in three phases. The first phase consisted of a simple scoping analysis that was performed to support the work group meeting. This was followed by a more in-depth analysis that was initiated after the St. Louis meeting (Phase 2). Finally, in light of the substantial uncertainties in the range of possible input parameters for the model (e.g., building volume, radon release fraction, and air exchange rate), we also performed a Monte Carlo simulation (Phase 3). Phases 2 and 3 are being reported herein to the work group for the first time.

4.1 Phase 1 – Scoping Analysis

Using the "Single Chamber Model" (Environmental Instruments Canada Inc. n/d), a scoping calculation was performed to estimate the range of potential radon levels in Blockson Building 40. This model calculates the radon concentration at equilibrium in a single enclosure using input values for the radon source term, the enclosure volume, and the ventilation rate. This model calculates the equilibrium radon concentration that results in an enclosed structure (box) for a given constant radon input rate (pCi/s) and a constant ventilation rate, expressed as building volume turnovers per hour (h^{-1}). For the purpose of this scoping analysis, we assumed that the radon in the phosphate rock was in full equilibrium with the ^{226}Ra at the time that the rock was transported into Building 40 for processing. The input rate of radon into the building atmosphere was assumed to be the product of the ^{226}Ra (Rn) activity concentration (pCi/ton), the radon release fraction (the fraction of radon atoms in the ore that escapes from the ore and enters the building volume), and the feed rate of the ore (tons/s). At equilibrium, the radon input rate is equal to the radon exit rate through ventilation and the radon concentration remains constant.

The radon source term was calculated using the "Radon Generated by a Process Model" (Environmental Instruments Canada Inc. n/d). This model calculates the radon source term using inputs of percent uranium in the ore, feed rate of the ore, and the release fraction for radon from the ore in the process. The values used were:

% U_3O_8: 0.014%
Ore Feed Rate: 35.7 tons/h (6,000 tons/week)
Release Fraction: 0.3[1]

This results in a calculated release rate of 4,370 Bq/s (1.18×10^5 pCi/s) of radon into Building 40. The volume of Building 40 was estimated, based on a drawing of the plant recently provided to SC&A by a former worker and a scale drawing by Wynveen et al. (1983), to be 24,000 m^3 (89 m × 28 m × 9.75 m). The "Single Chamber Model" was run for ventilation rates of 1, 2.5, and 5 h^{-1}. These ventilation rates were selected from the range of values for commercial buildings reported in several publications, primarily Fradella 2005, Parker 1985, and Turk 1989. In addition, a conversation with Dr. Mort Lippmann, Professor of Industrial Hygiene at New York University Medical Center, Institute of Environmental Medicine, revealed that in his opinion, the air turnover rate in a building such as Building 40 at Blockson would not be less than 1 per hour. Hence, we set 1 air turnover per hour as our lower bound in this scoping analysis.

Table 2 presents the results of scoping calculation that were performed by SC&A to assess the degree to which the radon concentrations measured at Florida facilities and the values measured

[1] In the analysis that was discussed at the St. Louis meeting, SC&A employed a default radon release fraction of 0.3, because it was believed to be the default radon emanation coefficient adopted in RESRAD. In preparing this report, we revisited this parameter and found that the current version of RESRAD uses a default radon emanation coefficient of 0.25.

in 1983 at Blockson are compatible and consistent with model results. Values similar to these[2] were presented and discussed at the work group meeting held in St. Louis on June 24, 2008.

Table 2. Phase 1 Parametric Analysis of Radon Concentrations in Building 40

Parameter	Ventilation Rate		
	1/hr	2.5/hr	5/hr
Radon (pCi/L)	17.5	7.1	3.5
WL[1]	0.07	0.028	0.014
WLM/y	.82	0.33	0.16

[1]Radon daughter equilibrium factor = 0.4 (See below)

For reference, the bounding radon assigned from OTIB-0043 was 2.33 pCi/L (0.112 WLM/y). SC&A's revised bounding radon value was 7.73 pCi/L (0.37 WLM/y). Hence, this scoping analysis revealed that the default radon concentration adopted by NIOSH in OTIB-0043 did not appear to be incompatible with the modeled results.[3]

The work group discussed this matter during the full Board meeting held on June 24–26, 2008. During that meeting, it became apparent that these scoping analyses were considered important by the work group and the Board. In addition, since the radon concentrations derived by SC&A and discussed at the meeting were not formally delivered to the work group and had not yet undergone SC&A's QA process, nor had the work group or NIOSH had a chance to review the reported values, the work group and the Board requested that SC&A complete its modeling evaluations, perform the required QA checks, and formally deliver its report on this matter to the work group as soon as possible. In response to this request, SC&A commenced additional follow-up investigations.

4.2 Phase 2 – Follow-up Investigations

Following the St. Louis meeting, SC&A initiated a more thorough review of the literature to gain further insight into the reliability of the model and the possible range of values for the model input parameters. Of particular importance is the radon source term in Building 40 and the air turnover rate. Table 3 shows the radon concentrations modeled for Building 40 using a full range of alternative assumptions for each of these parameters. A detailed description of that analysis is provided in Appendix B.

[2] The values presented and discussed during the work group meeting were lower than these, primarily due to revisions in our understanding of the volume of Building 40; i.e., subsequent to the St. Louis meeting, we determined that the footprint of Building 40 was smaller than that initially assumed and discussed during the St. Louis meeting.

[3] Also of interest is the relationship of radon concentration (at equilibrium) to ventilation rate. For a constant input rate of radon, the relationship of radon concentration to ventilation rate is inversely proportional; thus, a 5-fold increase in ventilation rate (1/hr to 5/hr) results in a 5-fold decrease in the equilibrium radon concentration. The working level will not be exactly proportional to the radon concentration, as the radon daughter equilibrium fraction decreases as the ventilation rate increases. However, if we adopt the typical indoor equilibrium fraction of 0.4 cited by UNSCEAR (2000), the inverse proportional relationship also applies to WL (and WLM/yr) and ventilation rate.

Table 3. Radon Concentrations (pCi/L)[b]

	Release fraction from acid (f)					
ACH[a]	**0.0**	**0.2**	**0.4**	**0.6**	**0.8**	**1.0**
0.5	0.44	18.52	36.61	54.70	72.79	90.88
1.5	0.15	6.24	12.33	18.42	24.51	30.60
2.5	0.09	3.75	7.41	11.07	14.73	18.39
3.5	0.06	2.68	5.30	7.92	10.53	13.15
4.5	0.05	2.09	4.12	6.16	8.20	10.23
5.5	0.04	1.71	3.37	5.04	6.71	8.37

[a] Air changes per hour

[b] A description of the models used to derive these values is provided in Appendix B.

As may be noted, depending on the modeling assumptions used, the average annual radon concentration in Building 40 during the qualification period could be as low as 0.04 pCi/L (which is well below typical outdoor natural background levels) to as high as 91 pCi/L. When confronted with the enormous uncertainty associated with the above "what if" analyses, SC&A perfomed a Monte Carlo analysis. Monte Carlo analyses are especially useful when deterministic analyses (i.e., those presented in Table 3) yield such variable results that they are of little use in making informed decisions.

4.3 Phase 3 – Monte Carlo Analysis

As described above, our follow-up investigations revealed that the input parameters used to derive the average annual radon concentration in Building 40 during the qualification period are highly uncertain, resulting in very different results, depending on the selected input parameters. In order to address these uncertainties in a more formal manner, we performed a Monte Carlo analysis. A detailed description of that analysis is provided in Appendix B. In summary, we assigned the following distributions to the key input parameters used in this calculation.

Table 4. Distributions of Input Parameters Used in Monte Carlo Analysis

Symbol	Description	Units	Distribution	Parm 1[a]	Parm 2[b]	Parm 3[c]	Fixed[d]
S	Specific activity of ^{226}Ra	Bq/kg	normal	1,460	287		1,460
t_r	residence time of ore in Building 40	s	triangular	7,200	14,400	21,600	14,400
ε_d	dry emanation coefficient	—	lognormal	0.113	1.545		0.113
ε_w	wet emanation coefficient	—	lognormal	0.299	1.498		0.299
f	evolution fraction of Rn from sulfuric acid	—	uniform	0.000	1.00		—
r	air exchange rate	s^{-1}	uniform	2.78e-05	1.53e-03		—
V	Volume of building	m^3	uniform	17,321	23,983		20,652

[a] Mean of normal distribution, geometric mean of lognormal, or lower limit of uniform or triangular distributions

[b] Standard deviation of normal distribution, geometric standard deviation of lognormal, upper limit of uniform distribution, or mode of triangular distribution

[c] Upper limit of triangular distribution

[d] Used in sensitivity analysis reported in Table 3

Note that the key parameters that contribute to the uncertainty are the following:

(1) Fraction of radon trapped in the crystalline structure of the pulverized phosphate rock that emanated from the reaction vessels and entered the Building 40 atmosphere

(2) Building air exchange rate

(3) Volume of building

Using these assigned distributions for the input parameters, we obtained the following results.

Table 5. Percentile Values of ^{222}Rn Concentrations (pCi/L)

Percentile	Value
0%	0.02
5%	0.78
10%	1.50
15%	2.24
20%	2.97
25%	3.70
30%	4.44
35%	5.20
40%	5.99
45%	6.84
50%	7.72
55%	8.73
60%	9.87
65%	11.28
70%	13.06
75%	15.54
80%	19.08
85%	24.77
90%	35.37
95%	61.95
100%	651.00

These results indicate that we can be 95% certain that the true but unknown average annual radon concentration in Building 40 during the qualification period was less than 62 pCi/L.

5.0 REFERENCES

Birky, B.K. et al. 1998. "Evaluation of Exposure to Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) in the Phosphate Industry: Final Report," FIPR 05-046-155 (Florida Institute of Phosphate Research). http://fipr1.state.fl.us/fipr/fipr1.nsf/9bb2fe8f45c4945e85256b58005abaec/deb248481f98842e85256b2f005d7524!OpenDocument

Birky, B.K. 2005. "Phosphate Plant Radon Levels," electronic mail to R. Gard (Oak Ridge Associated Universities Team), September 1, Florida Institute of Phosphate Research, Bartow, Florida.

Environmental Instruments Canada Inc. n/d. Uranium Safety Toolkit v2.0, , http://members.shaw.ca/eic/Tools/index.htm

EPA (U.S. Environmental Protection Agency) 1978. "Radiological Surveys Of Idaho Phosphate Ore Processing – The Wet Process Plant," Technical Note: ORP/LV–78–1, April 1978.

Fradella, J., and J. Siegel 2005. "An Evaluation of Shelter-In-Place Strategies in Four Industrial Building." Proceedings: Indoor Air 2005, pp. 3360–3362.

Guimond, R.J., W.A. Mills, S.T. Windham 1977. "Radiation Exposures in the Florida Phosphate Industry," International Radiation Protection Association Meeting #4, Paris, France, Volume 3, pp. 1049–1052.

Marseglia, L.M. 1983. "Joliet Radiation Survey." In Olin Corporation, Surveys and Radiological Data of the Former Blockson Chemical Company Site, provided to ORAUT, 2007 [SRDB Ref ID 30623].

OCAS (Office of Compensation Analysis and Support) 2007. Response to Remaining Issues for the Blockson SEC Evaluation Review by SC&A. Tom Tomes, National Institute for Occupational Safety and Health, OCAS. November 13, 2007.

ORAUT (Oak Ridge Associated Universities Team) 2006. *Characterization of Occupational Exposure to Radium and Radon Progeny During Recovery of Uranium from Phosphate Materials*, ORAUT-OTIB-0043, Rev. 00. January 6, 2006.

Parker, G. 1985. "Measurement of Air Exchange Rates in Residential and Commercial Buildings in the Northwest: Techniques and Results," PNL-SA-15607; CONF-8505167-1, 1985.

Slack, A.V. 1968. *Disposal or Use of Gypsum,* Phosphoric Acid, Vol. 1.

Tomes, T., and S. Glover 2007. *Technical Basis Document for Atomic Energy Operations at Blockson Chemical Company, Joliet, Illinois*," OCAS-TKBS-0002, Rev. 2. National Institute for Occupational Safety and Health, Office of Compensation Analysis and Support: http://www.cdc.gov/niosh/ocas/pdfs/tbd/ocblockson-r2.pdf

Turk, B., et.al, 1989. *Commercial Building Ventilation Rates and Particle Concentrations*, ASHREA Transactions, Vol.95, Part 1.

UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) 2000. "UNSCEAR 2000 Report to the General Assembly, with Scientific Annexes," Vol. I: "Sources," Annex B, "Exposures from Natural Radiation."
http://www.unscear.org/unscear/en/publications/2000_1.html

Wynveen, R. A., et al. 1983. "Formerly Utilized MED/AEC Sites Remedial Action Program: Radiological Survey of Chemicals Group, Olin Corporation (Formerly Blockson Chemical Company), Joliet, Illinois, March 27-November 28, 1978," DOE/EV-0005/35, ANL-OHS/HP-83-103. Argonne, Illinois: Argonne National Laboratory.

APPENDIX A: REVIEW OF RADON EXPOSURE ESTIMATES DERIVED FROM SUMMARY DATA IN ORAUT-OTIB-0043

A.1 Verification of OTIB-0043 Radon Exposure Estimates based on the Equal Weight Assumption

The summary data on radon exposures presented in Attachment B of ORAUT-OTIB-0043 (ORAUT 2006) were used by NIOSH to estimate the geometric mean (GM), geometric standard deviation (GSD), and 95th percentile of worker radon exposures. The OTIB calculations were performed with an assumption of equal weighting for all reported data points in the Attachment B tables that are not grayed out. The applicable data points include many values that are the means of sets of measurements, as well as many individual measurements. Some of the means represent large datasets, while other mean values represent smaller datasets.

The assumption of equal weighting is not applicable when the means represent very different sample sizes. However, the equal-weight assumption was used to develop the estimated radon exposures reported in Table 4-4 of OTIB-0043. In this section of the current report, the equal weight assumption is applied to confirm the NIOSH estimates. In the following section, the mean values in Table B-3 of OTIB-0043 are weighted proportionally to the sample size represented by each mean value to develop more appropriate weighted estimates of worker radon exposure.

Although OTIB-0043 states that 130 data values are contained in the non-shaded rows of the Attachment B tables, only 128 values are listed there, including seven values in Table B-1 that are below the limit of detection (LOD) reported as "<0.5 pCi/L." Two methods are used here to estimate a lognormal distribution using the equal-weight assumption: the maximum likelihood method (MLM) and a graphical method based on normal scores. The MLM requires that surrogate values be imputed for the seven LOD values. The midpoint of the uncertainty range (LOD/2 = 0.25 pCi/L) was used for these seven values. In the graphical method, these seven points are not included on the graph, but their ranks are used when computing the normal scores.

The results of these two analyses are compared with the OTIB-0043 estimates in Table A-1. As shown in the table, the lognormal distributions estimated using the MLM and graphical methods have a GM and GSD that are very similar to the GM and GSD reported in OTIB-0043 for the Attachment B data. The estimated (arithmetic) mean, standard deviation, and 95th percentile also closely match.

The normal score plot used in the graphical method is shown in Figure A-1. Note that the fitted regression line underestimates the high readings in the upper tail of the distribution. As shown in Table A-1, the 95th percentile of the empirical distribution (i.e., the data points in Figure A-1) is significantly higher than the 95th percentile estimated from the fitted lognormal distribution, 2.9 pCi/L versus the 2.3 pCi/L estimate reported in OTIB-0043.

A.2 Radon Exposure Estimates based on the Expanded Table B-3 Datasets

Many of the mean values reported in the Attachment B tables do not include detailed information on the underlying dataset. However, the mean values reported in Table B-3 of OTIB-0043 are accompanied by other statistics that describe the underlying dataset, such as the number of samples and the variance of the sample values. The sample sizes for these eight datasets range from 24 to 118, indicating that a wide range of weights should be considered. The smallest dataset in Table B-3 (labeled "gypsum stack flux test") has a mean value that is the second highest value contained in the entire dataset (6.52 pCi/L). Other, larger datasets summarized in Table B-3 also have relatively high mean values when compared to the mean value of approximately 1 pCi/L for the equally weighted data shown in Table A-1. Although other tables in Attachment B include mean values, only Table B-3 contains additional information on the variance of the underlying datasets from which the means were derived. The sample variance provides a measure of the spread of the values in each dataset about the mean. When the means are treated as single values, this information is lost. As a result, any estimate of the 95th percentile that does not include this information will be biased to the low side.

The mean values in Table B-3 were assigned weights in proportion to the size of sample each mean represents. All other valid data values in the Attachment B tables were assigned a weight of 1, as in the equally weighted approach. A weighted mean was calculated using these weights. The variance reported for each dataset in Table B-3 and the sample size were used to estimate the sum of squared deviations of the data values when determining the weighted standard deviation. The estimated weighted means and standard deviations were then used to calculate the parameters of a weighted lognormal distribution that represents the population of exposures in Attachment B. The required calculations are shown in Table A-2. The weighted mean is estimated as 1,482.49 ÷ 713 = 2.079 pCi/L, approximately twice as high as the unweighted mean value. The weighted standard deviation is estimated as $(17{,}834/713)^{1/2}$ = 5.01 pCi/L.

The weighted lognormal estimates are compared with the unweighted estimates from Section A-1 in Table A-3. Although both methods yield similar estimates for the GM, the weighted estimates have a GSD that is twice as large as the unweighted estimate. This results in a much higher estimate of the 95th percentile when the weighted approach is used (7.78 versus 2.33 pCi/L). A graphical comparison of the GM, GSD, and 95th percentiles is shown in Figure A-2.

Table A-1. Characteristics of Lognormal Distributions for Radon Measurements in OTIB-0043 Attachment B

Source of Estimates	Lognormal Parameters		N	GM (pCi/L)	GSD	95th Percentile (pCi/L)	Mean (pCi/L)	Standard deviation (pCi/L)
	mu	Sigma						
Maximum likelihood	-0.323	0.738	128	0.724	2.092	2.438	0.951	0.757
Graphical method	-0.351	0.784	128	0.704	2.191	2.558	0.958	0.790
OTIB-0043 estimates	-0.286	0.688	130	0.751	1.989	2.330	0.951	0.731
				Median				
Empirical Distribution	--	--	128	0.700	--	2.900	1.006	1.169

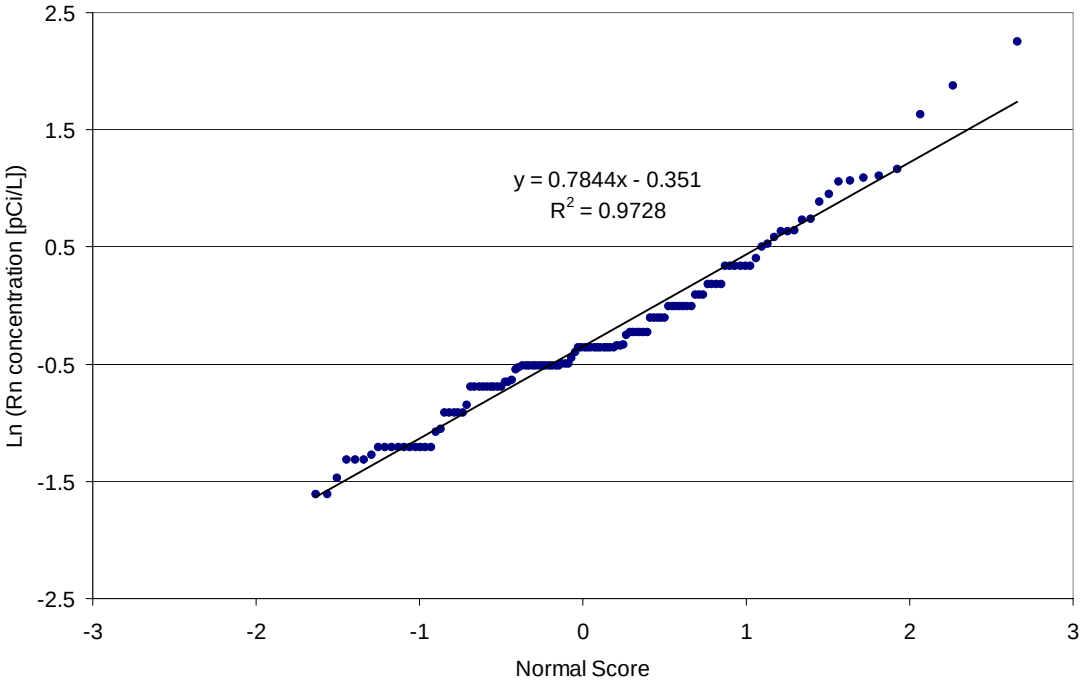

Figure A-1. Normal Score Plot of Attachment B Radon Measurements

Table A-2. Derivation of Weighted Lognormal Distribution Using Expanded Data for Table B-3 Means

Values for analysis:	Count (C)	Mean (μ)	Sample variance (V)	C · μ	C · V	Σ (x - μ)
NE gypsum stack well	90	2.43	18.45	218.7	1660.5	1671.57
Auto shop SE fence	56	2.89	23.73	161.84	1328.88	1365.69
SW of plant	31	0.35	0.22	10.85	6.82	99.52
Burn area fence	118	1.89	27.39	223.02	3232.02	3236.25
Liming station ladder	105	1.9	25.36	199.5	2662.8	2666.17
Environmental monitoring well	101	2.6	55.26	262.6	5581.26	5608.65
Gypsum stack flux test	24	6.52	24.52	156.48	588.48	1061.77
Cooling pond hand rail	68	2.08	26.85	141.44	1825.8	1825.80
All other Attachment B tables	120	0.9005		108.06	131.946	298.67
Total	713	2.079[a]		1482.49		17834.10

Note: Columns 1–4 from ORAUT (2006, Table B-3)

[a] μ_w = weighted mean

Table A-3. Comparison of Unweighted and Weighted Distributions for Attachment B Data

Lognormal models	μ_y [a]	σ_y [b]	GM (pCi/L)	GSD	95th percentile (pCi/L)
OTIB-0043	-0.286	0.688	0.75	1.99	2.33
Weighted Distribution	-0.226	1.384	0.80	3.99	7.78

[a] Mean of logs = ln(GM)

[b] Standard deviation of logs = ln(GSD)

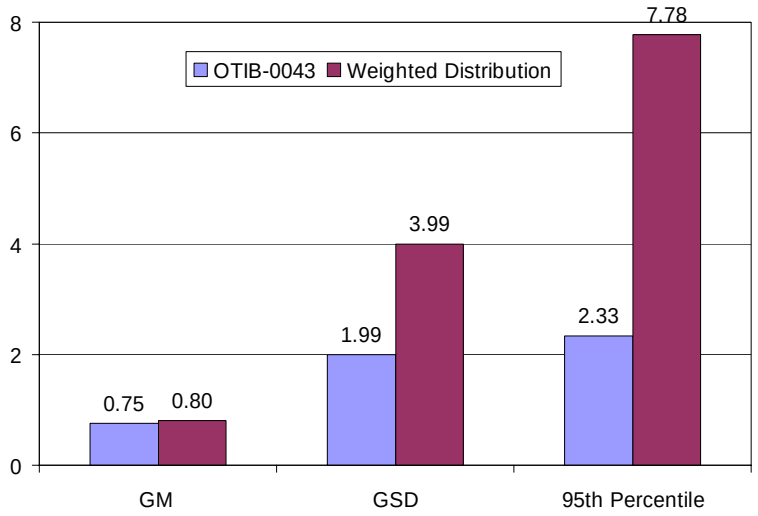

Figure A-2. Comparison of OTIB-0043 Estimates with Lognormal Parameter Estimates Using Expanded Data in Table B-3

APPENDIX B: COMPREHENSIVE ANALYSES OF ^{222}RN CONCENTRATIONS IN BUILDING 40

In order to gain greater insight into the possible range of radon concentrations in Building 40 during the qualification period, SC&A (1) performed a comprehensive review of available documentation and literature pertinent to modeling radon buildup inside structures in which phosphate rock was processed, (2) carefully reviewed Blockson documentation, and (3) interviewed several former Blockson workers. Using this information, we first performed two sets of analyses to estimate the average annual radon concentration in Building 40 during the qualification period. The first was a parametric analysis, in which we used the most likely values of several uncertain parameters, while varying stepwise the values of the two parameters most likely to affect the radon concentrations. In the second, we applied Monte Carlo techniques to construct a probability distribution of these concentrations.

We begin this discussion with an overview of the processes employed in Building 40 and of the characteristics of the building. We then present a detailed discussion of the analysis, including the sources of the data, and the results and conclusion.

B.1 Processes in Building 40

B.1.1 Processing of Phosphate Ore

According to "Emission Factor Documentation . . ." 1993:

> *Phosphate rock from the mines is first sent to beneficiation units to separate sand and clay and to remove impurities. Steps used in beneficiation depend on the type of rock. A typical beneficiation unit for separating phosphate rock mined in Florida begins with wet screening to separate pebble rock, which is larger than 1.43 millimeters (mm) (0.056 inch), or 14 mesh, and smaller than 6.35 mm (0.25 in.) from the balance of the rock. The pebble rock is shipped as pebble product. The material that is larger than 0.85 mm (0.033 in.), or 20 mesh, and smaller than 14 mesh is separated using hydrocyclones and finer mesh screens and is added to the pebble product. The fraction smaller than 20 mesh is treated by two-stage flotation. The flotation process uses hydrophilic or hydrophobic chemical reagents with aeration to separate suspended particles. Phosphate rock mined in North Carolina does not contain pebble rock. . . . Like Florida rock, the fraction that is less than 10 mesh is treated by two-stage flotation, and the fraction larger than 10 mesh is used for secondary road building.*

The ore received at the Blockson plant would have been beneficiated prior to shipment from Florida. The ore "is maintained at about 10 percent moisture" ("Emission Factor Documentation . . ." 1993). Twenty-mesh ore consists of particles < 0.841 mm in diameter (Perry and Green 1984). Very fine particles would have been removed by the floatation process. Once received at Blockson, the ore was stored in 100-ft-high silos until it was ready to be used for producing phosphoric acid. The first step in phosphate production was the calcining of the ore in an outdoor oven at temperatures of 1,400–1,600ºF (1,033–1,144 K). The calcined ore was then

transferred to Building 40 by means of a continuous-screw conveyer. "Dried or calcined rock is ground in roll or ball mills to a fine powder, typically specified as 60 percent by weight passing a 200-mesh sieve" ("Emission Factor Documentation . . ." 1993). (Two-hundred mesh corresponds to a particle diameter of 74 μm, Perry and Green 1984). The ground ore was then transferred to a tank of sulfuric acid. According to a former Blockson worker, 66% sulfuric acid was sent to Building 40, where it was further diluted with water. Since the dilution of sulfuric acid is an exothermic process, the temperature of the resulting liquid would be above ambient. According to another former worker, the tanks contained steam coils to heat the sulfuric acid further.

B.1.2 Emission of Radon

Radon-222 (referred to as "radon" in the remainder of this appendix) could have been released from the phosphate ore at various stages of the process. About 30% of the radon generated by the radioactive decay of ^{226}Ra in the wet ore would emanate into the pore space; the remaining 70% would remain in the crystalline matrix. During the calcining, virtually all volatile components, including water and radon, would be driven out of the rock. Some additional radon would grow in during the period of approximately 4 h during which the rock is transported through Building 40 and ground to the desired fineness. The radon emanation coefficient from the dry phosphate ore is approximately 11%.[4] Once the ore is transferred to the acid, the crystalline matrix would have been dissolved. Most of the ^{226}Ra would precipitate, but the radon would remain in solution. Since the warm acid was agitated during the digestion process, some of the dissolved radon could potentially have escaped into the air.

B.2 Characteristics of Building 40

B.2.1 Size of Building

The size of the interior of Building 40 plays an essential role in calculating the radon concentration in the ambient air: the larger the space, the lower the concentration. A former Blockson worker furnished SC&A and OCAS a plan of the Joliet plant and marked the location of Building 40 on this drawing (see Figure B-1). We used that drawing to identify Building 40 on a drawing of the plant showing soil sampling locations (Wynveen et al. 1983, Figure 10), which is reproduced in Figure B-2, with the location of Building 40 shown in red. We used the map scale displayed in the drawing to estimate the dimensions of Building 40. The interior is assumed to be 32 ft (9.75 m) high, the same as Building 55 (Lopker and Block 1951, Appendix B). According to a former worker, the building also housed a silica production operation, separated by a brick wall from the rest of the plant. This wall may be represented by a line shown in Figure B-1 that divides the north and south portions of the building, and the phosphoric acid operation may have taken place in the north part of the building.

[4] A more detailed discussion of these data is presented later in the present report.

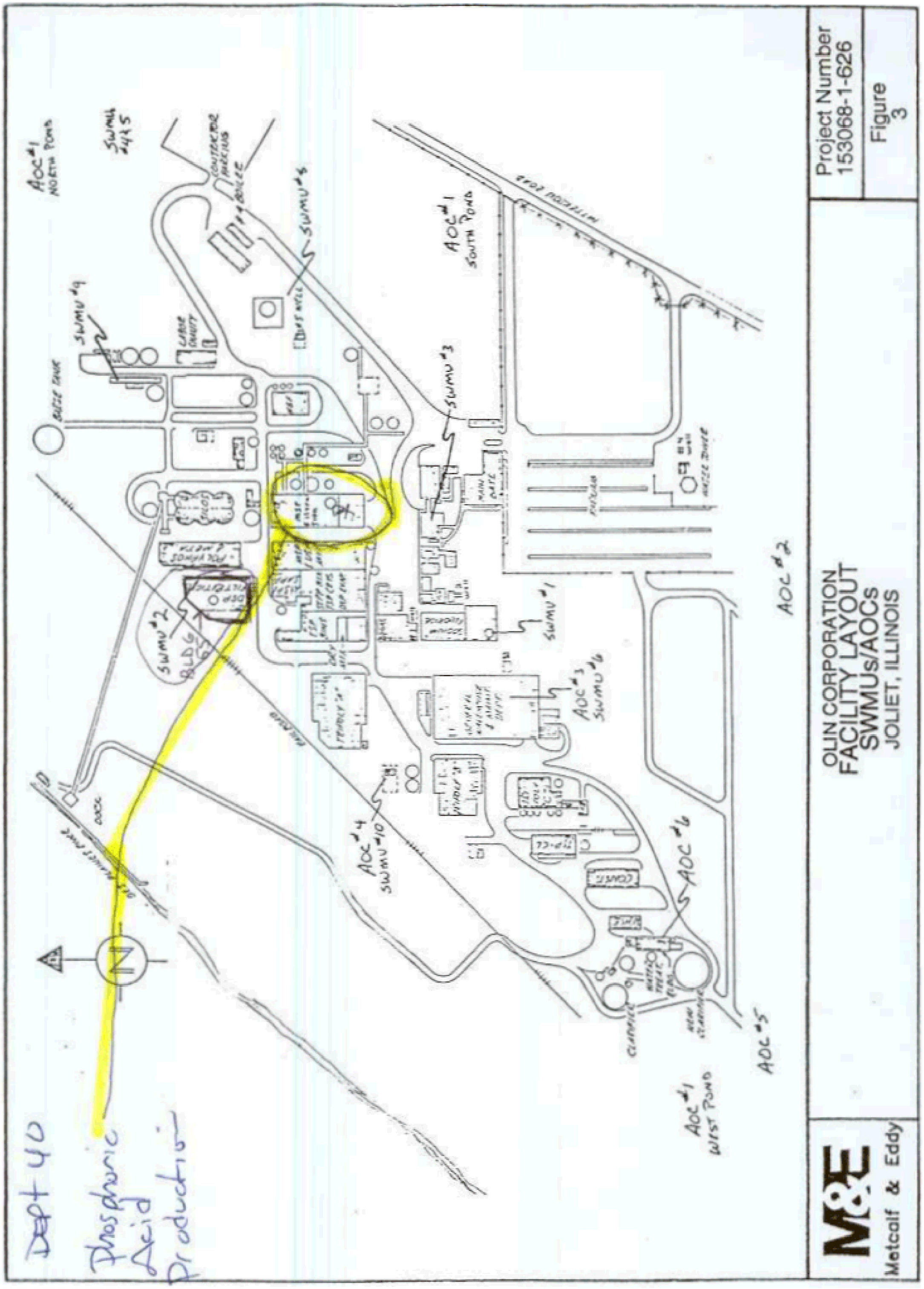

Figure B-1. Layout of Former Blockson Chemical Company Plant in Joliet, IL

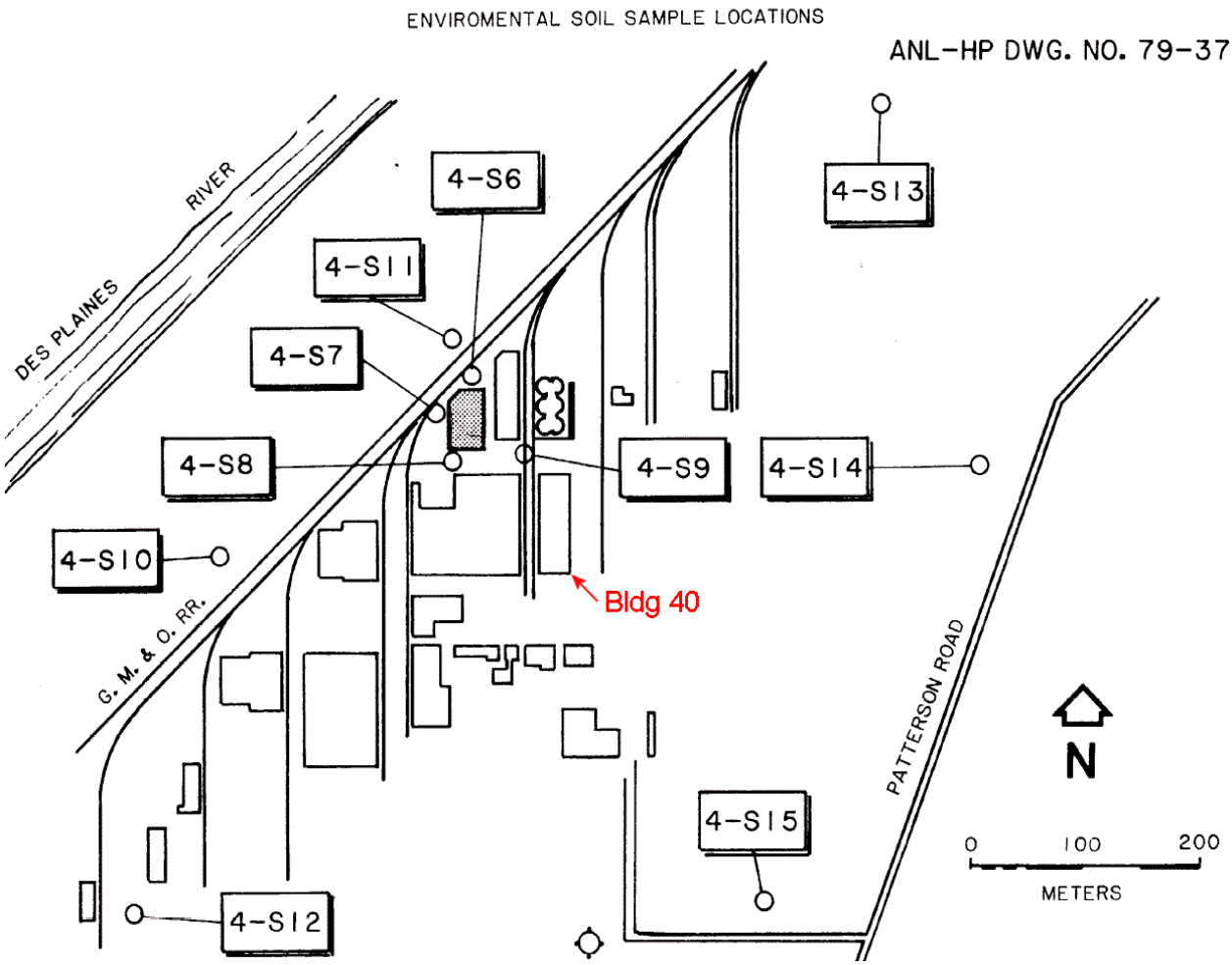

Figure B-2. Environmental Soil Sampling Locations at Former Blockson Chemical Company Plant (Wynveen et al. 1983, Figure 10)

B.2.2 Building Ventilation

The ventilation rate, also referred to as the air turnover rate, of Building 40 also plays a crucial role in calculating the radon concentrations. Three former workers were interviewed via teleconference by Wanda Munn, Chair of the Advisory Board Work Group on Blockson Chemical SEC; Tom Tomes of NIOSH/OCAS; and SC&A personnel. All three workers were asked about the building ventilation—their recollections varied. Only two of the workers had worked inside Building 40, so they would most likely have more accurate recollections about the conditions inside the building.

According to Worker #1 (the numbering is arbitrary):

> *The plant was generally a closed plant with just a few doors. There were small windows at the very top of the plant,*[5] *but he never saw them opened or could not say for sure if they could be opened. The few doors would be propped open during the hot summer months. There were a lot of fumes in the plant. . . . There was a stack that vented the tanks, but without forced air.* (Tomes 2008a)[6]

According to Worker #2:

> *The building had roof vents and roof fans. He said there were windows or openings on the first floor that allowed air to enter the building that could be pushed out, but were generally closed. He said they were on the long sides [of the rectangular building] and were about 3 feet by 5 feet. They supplied ventilation air for the digestors on the second floor.* (Tomes 2008b)[3]

Both workers stated that new ventilation was installed or existing ventilation upgraded some time between about 1960 and 1980.

B.3 Methodology

Radon could have been released into Building 40 through two mechanisms:

(1) Emanation from ore within the building, before, during, and after the grinding of the ore

(2) Evolution from the hot sulfuric acid after the crushed ore is dissolved

Radon emanates from the solid matrix of the phosphate ore by recoil of the radon atom following the emission of an alpha particle by its parent nuclide, ^{226}Ra. According to UNSCEAR (2000), "The range of recoil distance for ^{222}Rn is 20–70 nm in common minerals." This distance is less than 0.1% of the diameter of the crushed ore particles, which is less than 74 µm. Consequently, the crushing of the ore would have little effect on the radon emanation.

In mechanism 2, radon that has remained embedded in the solid matrix of the rock would enter into the acid solution after the rock has dissolved. There are no readily available data on the rate of evolution of radon from aqueous solutions. However, since the ~30% sulfuric acid is heated and agitated to promote the dissolution of the ore, such evolution cannot be ruled out. (See further discussion in Section B.5.)

The rate of release of radon into the building atmosphere, as well as its removal by air exchange and radioactive decay, is given by the following differential equation:

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[5] This observation is consistent with a photograph of Building 55 and an adjacent structure shown in Barr et al. 1953. Both buildings appear only to have windows just below the level of the roofs.

[6] Excerpted from interview notes prepared by Mr. Tomes—not a verbatim quote.

$$\frac{dB}{dt} = R\,S\left(t_r\,\varepsilon_d + \frac{f\left[\left(1-\varepsilon_d\right)\left(1-e^{-\lambda t_r}\right)+\left(1-\varepsilon_w\right)e^{-\lambda t_r}\right]}{\lambda}\right) - B\,(r+\lambda) \qquad (1)$$

$$= 0$$

B = atoms of daughter product (^{222}Rn)

R = processing rate of ore
= 9.45 kg/s
$= \frac{R_w}{t_w}$,

R_w = weekly processing rate
= 6,000 tons/week (Lopker and Block 1951) = 5.443×10^6 kg/week

t_w = weekly operating time
= 160 h/week = 576,000 s (assume three shifts/d, 7 d/week, 8 h downtime for maintenance)

S = specific activity of ^{226}Ra in phosphate rock (Bq/kg)

t_r = residence time of phosphate rock in Building 40 (s)

ε_d = emanation coefficient of radon from dry phosphate rock

f = fraction of radon evolving from sulfuric acid

ε_w = emanation coefficient of radon from wet phosphate rock

λ = decay rate of ^{222}Rn
= 2.0979×10^{-06} s^{-1}

r = outside air exchange rate inside Building 40 (s^{-1})

The first term inside the large parentheses in the first line of Equation 1 represents the rate at which radon emanates from the phosphate ore during its passage through Building 40. The second term (the algebraic fraction with λ in the denominator) represents the evolution of radon after the rock is dissolved in sulfuric acid. The first term in the numerator represents the evolution of radon that results from the decay of ^{226}Ra in the dry rock following calcining, while the second term represents the evolution of radon that was in secular equilibrium with ^{226}Ra in the wet rock prior to calcining. The final term of Equation 1 represents the removal of radon due to ventilation and radioactive decay. Radioactive decay of radon during the calcining step is not accounted for, nor is the reduction in the specific activity of ^{226}Ra due to radioactive decay during this process.

The second line in Equation 1 denotes the steady-state solution to the first line: the rate of change equals zero. This condition allows a direct solution of the first line in Equation 1:

$$B = \frac{R\,S\left(t_r\,\varepsilon_d + \frac{f\left[\left(1-\varepsilon_d\right)\left(1-e^{-\lambda t_r}\right)+\left(1-\varepsilon_w\right)e^{-\lambda t_r}\right]}{\lambda}\right)}{r+\lambda} \quad (2)$$

The activity concentration of radon in the ambient air is given by:

$$C = \frac{R\,S\left(t_r\,\varepsilon_d\,\lambda + f\left[\left(1-\varepsilon_d\right)\left(1-e^{-\lambda t_r}\right)+\left(1-\varepsilon_w\right)e^{-\lambda t_r}\right]\right)}{(r+\lambda)\,V} \quad (3)$$

C = activity concentration of radon in Building 40 (Bq/m^3)

V = volume of Building 40

B.3.1 Method of Calculation

Many of the parameters used to calculate the radon concentration, including the radon emanation coefficients, the specific activity of ^{226}Ra in phosphate ore, fraction of radon evolving from sulfuric acid, the air exchange rate, and the residence time of the ore, are variable and/or uncertain. In order to study the dependence of the radon concentrations on the values of the variable parameters, we performed two sets of analyses:

(1) Deterministic analysis, in which the parameters were assigned one or more fixed values (also referred to as "parametric analysis")

(2) Probabilistic analysis, using Monte Carlo methods

The variability and uncertainty in the values of these parameters can be expressed by probability distributions. For parameters that are uncertain or variable, Monte Carlo sampling methods are used to pick the particular set of values in a given calculation, called a realization. In the present analysis, the estimation of each concentration involved 100,000 realizations. Each individual calculation is deterministic—it employs a set of fixed values of the relevant parameters and produces a fixed result. These 100,000 results themselves form a probability distribution. Thus, the radon concentration can be expressed in terms of a mean, a standard deviation, the median, or, say, the 95th percentile. The analysis was performed using the Microsoft Excel add-in, Crystal Ball Version 2000.2 (Decisioneering 2001).

B.3.2 Input Parameters

In the following sections, we discuss the values of the uncertain parameters. The distributions used in the uncertainty analysis are discussed first, followed by the fixed value or range of values selected

B.3.2.1 Specific Activity of ^{226}Ra in Phosphate Rock

Hull and Burnett (1996) reported the specific activities of ^{226}Ra in 13 samples of phosphate rock from central Florida.[7] Only the mean and standard deviation of the individual assays are listed in their report. In the present analysis, the specific activity of ^{226}Ra is represented by a normal distribution, with the same mean and standard deviation as reported by Hull and Burnett. The parameters characterizing this distribution are listed in Table B-1. The mean (listed in the last column) is used as a fixed value in the parametric analysis.

Table B-1. Distributions of Input Parameters Used in Monte Carlo Analysis

Symbol	Description	Units	Distribution	Parm 1[a]	Parm 2[b]	Parm 3[c]	Fixed[d]
S	specific activity of ^{226}Ra	Bq/kg	normal	1,460	287		1,460
t_r	residence time of ore in Building 40	s	triangular	7,200	14,400	21,600	14,400
ε_d	dry emanation coefficient	—	lognormal	0.113	1.545		0.113
ε_w	wet emanation coefficient	—	lognormal	0.299	1.498		0.299
f	evolution fraction of Rn from sulfuric acid	—	uniform	0.000	1.00		—
r	air exchange rate	s^{-1}	uniform	2.78e-05	1.53e-03		—
V	Volume of building	m^3	uniform	17,321	23,983		20,652

[a] Mean of normal distribution, geometric mean of lognormal, or lower limit of uniform or triangular distributions.

[b] Standard deviation of normal distribution, geometric standard deviation of lognormal, upper limit of uniform distribution, or mode of triangular distribution.

[c] Upper limit of triangular distribution.

[d] Fixed values used in parametric analysis

B.3.2.2 Residence Time of Phosphate Ore in Building 40

We estimate that the time required to grind the ore, together with the transfer time to the grinder and then to the acid tank, to be 4 h, with a range of 2 to 6 h. The residence time is therefore represented by a triangular distribution with the aforementioned parameters. The parameters used in the calculation are listed in Table B-1. The mode is used as a fixed value in the parametric analysis.

B.3.2.3 Emanation Coefficients of Radon

Burnett et al. (1988) reported the loss of radon from samples of bulk phosphate rock under both wet and dry conditions. Ten of the samples were from Florida phosphate deposits. Because the data resemble a lognormal more than a normal distribution, we calculated the geometric mean and the geometric standard deviation for each set of measurements. Because one measurement of the dry emanation coefficient appeared to be an outlier, we utilized only nine of the measured values. We represented both the wet and the dry emanation coefficients by lognormal distributions. The geometric mean and the geometric deviation of each set of measurements are listed in Table B-1, which were adopted for the distributions in the present analysis. The geometric mean is used as a fixed value in the parametric analysis.

[7] The phosphate from this region has higher concentrations of radionuclides than the rock from northern Florida (the other region studied by these authors) and is therefore adopted to produce a claimant-favorable analysis.

B.3.2.4 Fraction of Radon Evolving from Sulfuric Acid

In the absence of available data on the evolution of radon from dilute, hot sulfuric acid, the fraction of evolved radon was assigned a uniform distribution ranging from 0 to 1. This parameter is varied stepwise over the same range in the parametric analysis.

B.3.2.5 Air Exchange Rate in Building 40

Sparse data exist on the exchange rates of outside air for industrial buildings, such as Building 40. The recollection of two former workers who worked inside the building differ as to the placement of the windows and the existence of forced air exhaust systems during the covered period.

The most applicable data was presented by Parker (1985), who measured the air exchange rates in two industrial buildings, using SF_6 as a tracer. One building was a machine/wood shop, with a floor area of 20,000 ft^2 (~1,860 m^2). This is comparable to the area of Building 40, which is estimated to be 1,776 m^2. General ventilation of the shop is through 12-ft × 25-ft bay doors, which were opened periodically during the 2-h period of measurement. The shop does not have a central HVAC system—local ventilation is provided for welding or metal stripping. The measured rate was 5.5 air changes per hour. Three measurements made when the shop was unoccupied and all outside doors were closed showed an average rate of 0.1 air changes per hour.

A second building studied by Parker was a single-story warehouse of approximately 100,000 ft^2 (~9,300 m^2), over five times the area of Building 40. According to Parker:

> *The warehouse is heated by overhead forced air electric heaters, but has no central HVAC system. Cooling is via several roof-mounted mechanically opened vents. The warehouse contains several 12-ft × 25-ft bay doors at either end of the building. During a working day, these doors are normally open and one or two receiving doors on the west side of the building are opened periodically.*

Measured rates during a working day were 0.05 air changes per hour in the morning and 0.2 in the afternoon. The rates averaged about 0.06 during non-working hours.

Other data primarily apply to residential structures. Yu et al. (1993) list a default rate of 0.5 air changes per hour for use with the RESRAD computer code. EPA (1997, Table 17-31) recommends a median of 0.45 air changes per hour for risk assessment studies. Godish (2001, Figure 11.5) indicates that ~44% of North American houses had rates of 0.25–0.5 air changes per hour in the early 1980's.

We assigned a uniform distribution of 0.1–5.5 air changes per hour to the ventilation rate of Building 40. The low end of the distribution is based on the geometric mean of two measurements during working hours in the warehouse reported by Parker (1985). The high end is based on the single measurement during working hours in the machine/wood shop reported by the same author. The values used in the analysis are listed in Table B-1. This parameter was varied stepwise over a range of 0.5–5.5 air changes per hour in the parametric analysis.

B.4 Results

The results of the parametric analysis of radon concentrations were presented in Table 3 of the main body of the present report. A statistical summary of the probabilistic analysis is presented in Table B-2. The percentile values are listed in Table B-3, while Figure B-3 is a graphical representation of the probability distribution. This distribution has a mean of 16.6 ± 0.1 pCi/L, a median of 7.72 pCi/L, and a 95th percentile of 61.95 pCi/L. Judging from Figure B-3 and from the percentile values, the distribution is approximately lognormal, with a geometric standard deviation of 3.55, as calculated from the median and the 95th percentile values.

Table B-2. Statistical Summary of Monte Carlo Analysis: ^{222}Rn Concentrations (pCi/L)

Statistic	Value
Trials	99,861
Mean	16.63
Median	7.72
Standard Deviation	31.76
Variance	1,008.41
Skewness	5.88
Kurtosis	52.35
Coeff. of Variability	1.91
Range Minimum	0.02
Range Maximum	651.00
Range Width	650.98
Mean Std. Error	0.10

Table B-3. Percentile Values of ^{222}Rn Concentrations (pCi/L)

Percentile	Value
0%	0.02
5%	0.78
10%	1.50
15%	2.24
20%	2.97
25%	3.70
30%	4.44
35%	5.20
40%	5.99
45%	6.84
50%	7.72
55%	8.73
60%	9.87
65%	11.28
70%	13.06
75%	15.54
80%	19.08
85%	24.77
90%	35.37
95%	61.95
100%	651.00

B.5 Discussion of Probabilistic Analysis

The distribution of radon concentrations spans a wide range, with a 70-fold difference between the 5th and 95th percentile values, indicating a large uncertainty in the analysis. As was discussed in Section B.3, the analysis incorporates a number of uncertain input parameters. The greatest uncertainty involves the fraction of the radon in the matrix of the phosphate rock that evolves from the aqueous sulfuric acid before the acid is pumped out of Building 40. Radon is quite soluble in cold water. In equilibrium, the volumetric concentration of radon in water at 0ºC is 51% of its concentration in the gas phase. However, that ratio is reduced to 11.4% at 50ºC and to 8.6% at 10ºC (Jenkins and Cook 1961, Table II). Thus, even for water at 0ºC, the equilibrium concentration in the air would be about twice that in the water. Given the much larger volume of building air, which is constantly being exchanged with the outside air, virtually all the radon would evolve into the atmosphere under static equilibrium conditions (i.e., no influx or removal of the aqueous sulfuric acid). An even greater fraction of the radon would evolve since, as stated earlier, the acid was kept at an elevated temperature. However, these observations do not cast any light on the *rate* at which the radon evolves from the acid. Since, according to our model, much more radon is dissolved in the acid than evolves from the ore in the building before it is dissolved in the acid, this uncertainty can have a profound effect on the results.

A lesser but still significant source of uncertainty is the building air exchange rate. The upper end of the range of this parameter is over 50 times greater than the lower end. According to Equation 3, the radon concentration is inversely proportional to $r + \lambda$, the air exchange rate plus the radioactive decay constant of ^{222}Rn. However, since the ventilation rate (0.1–5.5 air changes per hour), is far greater than the decay constant (~0.00755 h^{-1}), the concentration essentially varies as the reciprocal of the ventilation rate. The ventilation rate can thus cause an approximately 50-fold difference in the radon concentration. A still lesser source of uncertainty, not explicitly addressed by our model, is the possibility of incomplete mixing of the building atmosphere with the outside air. Such incomplete mixing would most likely have a lesser effect on the local radon concentration than the uncertainty in the ventilation rate—this effect is most likely subsumed by the wide range of ventilation rates employed in the analysis.

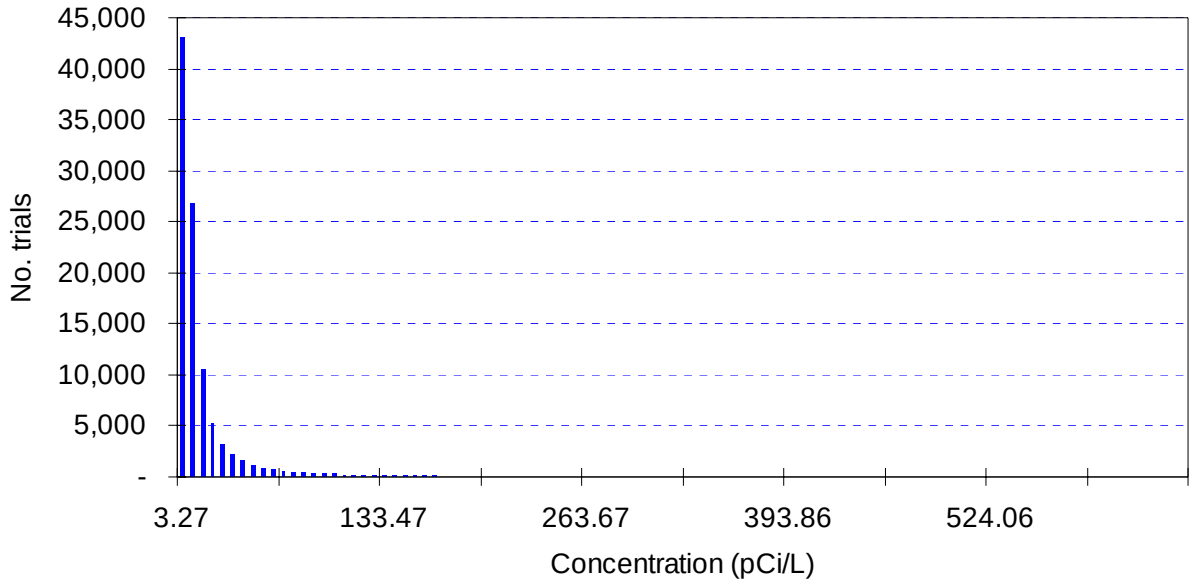

Figure B-3. Probability Distribution of ^{222}Rn Concentrations (pCi/L)

B.6 Conclusion

We conclude that the results of this analysis can be used to place a plausible upper bound on the average radon concentrations in Building 40. For instance, it is quite unlikely that the average concentration would have exceeded 62 pCi/L, the 95th percentile value of our probabilistic analysis.

Appendix B References

Barr, J.A., Jr., J.W. Ruch, and R.F. Borlik. 1955. "Recovering Uranium As By-Product in Phosphate Processing," *Rock Products*, (October), 96–102.

Burnett, W.C., et al. 1988. "Release of Radium and Other Decay-Series Isotopes From Florida Phosphate Rock," FIPR 05-016-059 (Florida Institute of Phosphate Research). http://www1.fipr.state.fl.us/FIPR/FIPR1.nsf/659556957d393fd685256afc000a4af1/31573005bc536bd885256b2f005ad314!OpenDocument.

Decisioneering, Inc., 2001. Crystal Ball 2000 (Version 2000.2) [Computer software and manual]. Denver, Colorado: Author.

"Emission Factor Documentation for AP-42 section 8.18 (now 11.21), Phosphate Rock Processing." 1993. Washington, DC: U.S. Environmental Protection Agency. http://www.epa.gov/ttn/chief/ap42/ch11/bgdocs/b11s21.pdf.

EPA (U.S. Environmental Protection Agency) 1997. "Exposure Factors Handbook." EPA/600/P-95/002Fa. Washington, DC: Author.

Godish, T. 2001. *Indoor Environmental Quality*. Boca Raton, Florida: Lewis Publishers.

Hull, C.D., and W.C. Burnett, 1996. "Radiochemistry of Florida Phosphogypsum." *Journal of Environmental Radioactivity, 32*(3), 213-238.

Jenkins, A.C., and G.H. Cook, 1961. "Chapter VII: Gas Phase Properties." In G. H. Cook (Ed.), *Argon, Helium and the Rare Gases: Vol. 1. History, Occurrence, and Properties.* New York: Interscience Publishers.

Lopker, E.B., and L. Block, 1951. Letter from E.B. Lopker and L. Block (Blockson Chemical Company) to Sheldon Wimpfen, U. S. Atomic Energy Commission. July 31, 1951.

Parker, G.B. 1985. "Measurement of Air Exchange Rates in Residential and Commercial Buildings in the Northwest: Techniques and Results," PNL-SA-13057, DE85012444.

Perry, R.H., and D.W. Green, 1984. *Perry's Chemical Engineers' Handbook* (6th ed.) New York: McGraw-Hill Book Company.

Tomes, T., 2008a. "Summary of Interview with [name redacted] Conducted by Phone on June 16, 2008, from approximately 12:00 PM until 12:40 PM."

Tomes, T., 2008b. "Summary of Interview with [name redacted] Conducted by Phone on June 17, 2008, from approximately 2:15 PM until 3:00 PM."

UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) 2000. "UNSCEAR 2000 Report to the General Assembly, with Scientific Annexes," Vol. I: "Sources," Annex B, "Exposures from Natural Radiation." http://www.unscear.org/unscear/en/publications/2000_1.html

Wynveen, R.A., et al. 1983. "Formerly Utilized MED/AEC Sites Remedial Action Program: Radiological Survey of Chemicals Group, Olin Corporation (Formerly Blockson Chemical Company), Joliet, Illinois, March 27–November 28, 1978," DOE/EV-0005/35, ANL-OHS/HP-83-103. Argonne, Illinois: Argonne National Laboratory.

Yu, C. et al., 1993. "Data Collection Handbook to Support Modeling the Impacts of Radioactive Materials in Soil," ANL/EAIS-8. Argonne, Illinois: Argonne National Laboratory.